Gas/liquid mass transfer in carbon dioxide–alkanes mixtures

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Abstract

The present paper tries to characterize the gas/liquid mass transfer processes in relation to the absorbent phase, employing binary mixtures of normal alkanes as liquid phase. The absorbent phase was in each case, had different binary relations between dodecane + heptane and dodecane + octane. Firstly, the absorption process in pure liquid phase was evaluated and characterized as regards to the operational variables. The effects produced by the mixture composition and the stirring rate upon the mass transfer process were also studied, in relation to the use of binary mixtures as liquid phases.
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1. Introduction

Mass transfer processes have centred an important part of the world research in the last decades. Research fields have been based on equipment design, scale-up, hydrodynamic behaviour, effects due to substances or contaminants presence, prediction, etc. And they have been the aim of numerous studies, which have produced several research papers [1–4]. The trend observed in the last years in mass transfer studies, being involved gas and liquid phases, has derived to the presence of organic compounds in the phases involved in absorption processes [5,6]. Removed strategies of these kinds of compounds from gas or liquid phases have caused the development of technical methodologies previously published [7,8]. For this reason, the presence of these kinds of substances in mass transfer operations is quite common, as different industries use these compounds as reagents, products, by-products or wastes.

Several authors have developed studies of oxygen mass transfer to different organic phases [9] such as toluene, decalin, ethanol, etc. The objective of these studies was characterizing the absorption processes of different gases (air, carbon dioxide, helium or hydrogen) in organic phases, on the basis of gas density and the gas hold-up.

This interest was begun by Linek and Benes [10] on the basis of the study of absorption processes employing emulsions of water in oil as liquid phase. These phases are complex. Their use as absorbent phases to remove or absorb gases implies a deep study of the absorption process and the effect upon the gas mass transfer kinetics of different typical operational variables, such as temperature, stirring rate and gas flow rate, as well as the composition of these liquid phases. Emulsions and microemulsions [7,11] are examples of these kinds of complex liquid phases used nowadays. The experimental data obtained by these researchers allow the conclusion that there is not an influence of liquid phase composition upon the mass transfer coefficient value in the systems studied. This behaviour was assigned to the ease the alkane presents in the gas phase to be absorbed to the organic liquid phase present in the emulsion.

Nowadays, other researchers have developed studies involved in gas/liquid/liquid systems, where different chemical reactions have been carried out in the aqueous phase, as well as analysing the global mass transfer process upon the value of the kinetics' constants [8].

Our research group has also developed also previous studies in relation to gas absorption in complex liquid phases, as Linek and Benes [10] and Cents et al. [8] have done. To be more specific, we used water in oil microemulsions (with an alkane as continuous phase) as the absorbent phase [12].

All studies previously mentioned show the interest in analysing the influence produced by the presence of organic substances in the liquid phases on mass transfer processes. In

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Nomenclature

A	interfacial area ($m^2\,m^{-3}$)
C	carbon dioxide concentration ($mol\,dm^{-3}$)
C_1	constant as defined in Eq. (8)
C_2	constant as defined in Eq. (10) ($cm\,s^{-1}$)
C^*	carbon dioxide solubility ($mol\,dm^{-3}$)
D	stirrer diameter (m)
D	gas diffusivity ($cm^2\,s^{-1}$)
F	constant as defined in Eq. (6)
K	constant as defined in Eq. (1) ($mm^2\,s^{-2}$)
K	constant as defined in Eq. (5) (s^{-1})
k_L	liquid side mass transfer coefficient ($cm\,s^{-1}$)
N	stirring rate (s^{-1})
P_{mo}	power for unaerated stirred vessel ($W\,m^{-3}$)
M	constant as defined in Eq. (7) ($cm^2\,s^{-1}\cdot(mPa\,s)^{-q}$)
M	constant as defined in Eq. (7)
Q_g	gas flow rate ($m^3\,s^{-1}$)
t	operation time (s)
V	vessel volume (m^3)
W	stirring rate (rpm)

Greek symbols

η	liquid viscosity (mPa s)
θ	time correction (s)
ν	kinematic viscosity ($mm^2\,s^{-1}$)
ρ	density ($g\,cm^{-3}$)

Numbers

Re	Reynolds number
Sc	Schmidt number
Sh	Sherwood number

this paper, we evaluate the behaviour observed by the use of different alkanes as absorbent phases in the mass transfer process of carbon dioxide from a gas to a liquid phase.

2. Experimental

The reagents employed in the present paper (dodecane, octane and heptane) have been supplied by Aldrich and Fluka with a purity of >99% for the octane and heptane and >97% for the dodecane.

Since binary mixtures have been employed in the present study attending to the research in the physicochemical characterization of multicomponent mixtures, it is necessary to take into account that there are clear deviations in relation to the linear behaviour. For this reason, the characterization in relation to the value of density and kinematic viscosity, those are important properties in mass transfer processes, was carried out and compared to the bibliographic data. Bi-distilled water and pure components of the mixtures were employed to prove that the methods employed were the adequate.

The kinematic viscosity (ν) was determined from the transit time of the liquid meniscus through a capillary viscometer

supplied by Schott (Cap No. 0c, 0.46 ± 0.01 mm of internal diameter, $K = 0.003201\,mm^2\,s^{-2}$) using Eq. (1). The viscometer used was a Schott-Geräte AVS 350 Ubbelohde type

$$\nu = K(t - \theta) \tag{1}$$

where t is the transit time, K the characteristic constant of the capillary viscosimeter and θ is a correction value to prevent the final effects. An electronic stopwatch with a precision of ± 0.01 s was used for measuring times.

The glass capillary was immersed in a bath controlled to $\pm 0.1\,^\circ C$ to determine the viscosity at $25\,^\circ C$. Each measurement was repeated at least six times. The dynamic viscosity (η) could be obtained by the product of kinematic viscosity (ν) and the corresponding density (ρ) of the binary mixture, in terms of Eq. (2) for each temperature and mixture composition.

$$\eta = \rho \cdot \nu \tag{2}$$

Density measurements of the corresponding mixtures were carried out using pycnometers (Gay-Lussac's pycnometer with a bulb volume of $25\,cm^3$). The pycnometers were placed into a thermostatic bath maintained at a constant temperature of $\pm 0.1\,^\circ C$.

Mass transfer studies of carbon dioxide to liquid alkanes were carried out using the experimental set up employed in previous works related to absorption processes, described in detail in a previous paper [13]. The gas/liquid contactor has been a 1-L volume cylindrical flat stirred vessel made of glass (internal diameter = 12 cm; height = 22 cm). Four baffles were placed on its internal wall from top to bottom in order to improve the mixing and prevent vortex formation during the experiment. The absorption processes has been carried out at $25\,^\circ C$ using a thermostat with a precision of $\pm 0.1\,^\circ C$. A six-blade Rushton turbine was used to stir the liquid phase. The gas flow rate was measured and controlled with two mass flow controllers (5850 Brooks Instruments). The apparatus employed in the present study to measure the gas flow rate and pressure was calibrated by the supplier.

The present paper analyses the effect caused by the stirring rate upon the mass transfer process kinetics of carbon dioxide to organic liquid phases (alkanes). In this study, several organic compounds (heptane, octane and dodecane) and its mixtures have been employed as liquid phases.

The stirring rate used in the absorption experiments was changed between 50 and 450 rpm. A constant value of $7.5\,L\,h^{-1}$ was employed in relation to gas flow rate.

3. Results and discussion

3.1. Physical properties

To analyse the gas/liquid mass transfer process it is necessary to take into account several factors that could have an influence in appreciating the grade in the process developed. Specifically, different physical properties must be analysed when mass transfer operations are being studied [14,15]. Viscosity is a property with a great influence in numerous chemical engineering processes. In

the present paper, the value of the density and kinematic viscosity were determined for the mixtures employed. The experimental results obtained for these physical properties were compared with the values obtained from literature and they agree with the data previously published [16]. The behaviour observed shows an increase in the viscosity value when the dodecane concentration in the mixture increases too. Also, the density shows a similar increase. The study of the data obtained for these two properties indicates that there are clear deviations in the mixtures employed in the present paper as regards the linear behaviour. Negative deviations were found for viscosity, while density exhibits positive ones.

3.2. Absorption process

Taking into account the operation regime employed in the present paper, the mathematical expression that governs the absorption process is shown in Eq. (3), obtained using mass balances to the gas phase due to the fact that the liquid phase was employed in batch regime.

$$\frac{dC}{dt} = k_L \cdot a \cdot (C^* - C) \tag{3}$$

where C^* is the gas solubility in the operation conditions, C the instantaneous gas concentration in the liquid phase, and $k_L \cdot a$ the liquid side volumetric mass transfer coefficient that results from the product of mass transfer coefficient and the mass transfer specific area. Eq. (3) is integrated along the operation time and adopts the expression shown in Eq. (4).

$$\ln\left(\frac{C^*}{C^* - C}\right) = k_L \cdot a \cdot t \tag{4}$$

In order to use Eqs. (3) and (4) it is necessary to know the value of the gas solubility in the liquid phase employed as absorbent, and then, to calculate the volumetric mass transfer coefficient. The solubility of carbon dioxide employed in the liquid phases was calculated using Eq. (5), thereby determining the gas concentration value at infinite time. These data agree (for linear alkanes) with the results obtained from reference [17] with discrepancies fewer than ±5%. The good results for solubility data obtained from absorption experiments indicate that experimental set-up allows the calculation of the mass transfer coefficient with acceptable results. When the chain length increases, a clear decrease is produced in the solubility value.

$$C = C^* \cdot (1 - e^{-k \cdot t}) \tag{5}$$

The solubility data have been employed to calculate de volumetric mass transfer coefficients by means of the ln graphical representation $(C^*/(C^* - C))$ versus operation time, and with acceptable linear regressions.

The gas/liquid interfacial area was obtained through contact device geometry, with a $\approx 109\,cm^2$ value. So the mass transfer coefficient corresponding to the liquid side was calculated using this area value.

On the basis of the contact device employed in the present paper (constant interface area), the effect of the gas flow rate

must be negligible. However, we had already observed in a previous paper that using this gas/liquid contactor [18] there is a certain influence of this variable upon the mass transfer operation, although in a lesser degree than the effect caused by the stirring rate. This influence due to the gas flow rate upon the mass transfer kinetics is due to the way of feeding the contactor with the gas; in this case, it is a direct jet on the liquid surface. It produces ripples at the mass transfer interface, with a higher effect when the gas flow rate is bigger. The importance of this effect is reduced when the viscosity of the liquid phase is increased (when the alkane increases the number of carbons), because it reduces modifications on the liquid surface.

Experimental results commonly indicate that both variables cause a power trend effect [19,20]. The stirring rate of the mechanical stirrer produces more effect than the feed gas flow rate. The power supplied to the liquid phase is commonly employed at any rate in the mass transfer and hydrodynamic studies in a mechanical stirred vessel. The values for the power supplied are obtained using Eq. (6) [20],

$$P_{mo} = \frac{f \cdot N^3 \cdot d^5 \cdot \rho}{V} \tag{6}$$

where N is the stirring rate, d the diameter of the stirrer, ρ the density of the stirred liquid, V the liquid phase volume and f is the graphically obtained factor that depends on the kind and characteristic length of the stirrer. For a Rushton turbine stirred f has a constant value equal to 1.8 [21].

3.2.1. Pure alkanes

Fig. 1 shows a typical example of the absorption kinetics obtained from the analysed cases in the present study. The liquid phase increases the gas concentration (carbon dioxide) along the operation time until the gas concentration in the liquid phase reaches a constant value. This value is the solubility data for this gas in the alkane at atmospheric pressure and 25 °C. The linear alkanes employed in the present paper as liquid phase have been previously analysed and their results published by our research group [18].
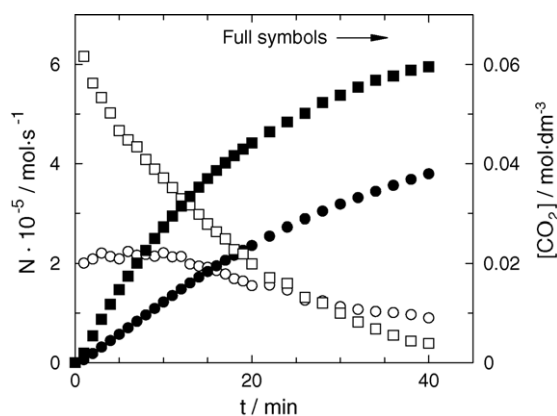

Fig. 1. Carbon dioxide absorption kinetics in alkanes mixtures at different values of stirring rate and $Q_g = 7.5\,L\,h^{-1}$. (O) and (●) for experiments at $W = 50$ rpm; (□) and (■) for experiments at $W = 450$ rpm.
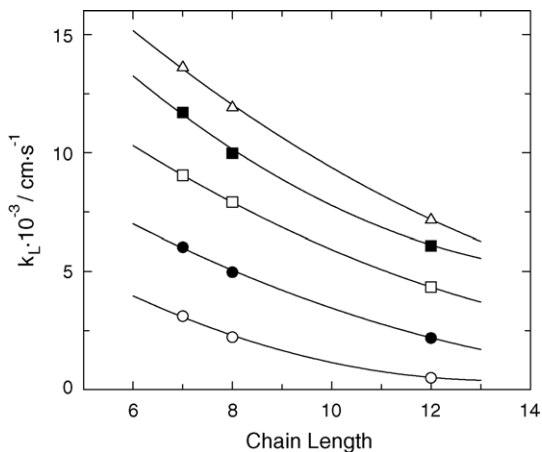
Fig. 2. Influence of chain length upon the mass transfer coefficients at different stirring rate values. (O) $W = 50$ rpm; (●) $W = 150$ rpm; (□) $W = 250$ rpm; (■) $W = 350$ rpm; (△) $W = 450$ rpm; $Q_g = 7.5$ L h^{-1}.

In Fig. 1, two experiments have been compared. The same liquid phase was employed in both experiments, while the stirring rate was changed. The experimental data indicates that a higher value in stirring rate produces that the saturation of the liquid phase will be reached in a minor operation time. Similar trends have been found when it was analysed that the effect of stirring power supplied to the liquid phase and the gas flow rate feed upon the gas/liquid mass transfer process.

An increase in the mass transfer velocity was observed in all pure alkanes employed in the present paper when stirring rate and gas flow rate was also increased. The enhancement in the gas/liquid mass transfer decreased when these variables reached high values (in the studied ranges) [18].

A comparison of experimental results is shown in Fig. 2. These results were obtained from the alkanes studied in the present paper, on the basis of liquid side mass transfer coefficient, where it is possible to observe that when the chain length increases, the velocity of gas/liquid mass transfer decreases due to the increase of viscosity in the liquid phase (Table 1).

Table 1
Physical properties for the alkanes employed and its mixtures at 25 °C

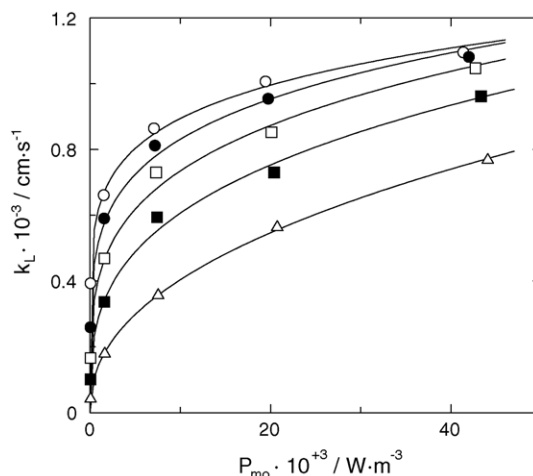| Liquid phase	ρ (g cm^{-3})	ν (mm^2 s^{-1})	$D \cdot 10^5$ (cm^2 s^{-1})
Heptane (C$_7$)	0.6795	0.3865	5.80
Octane (C$_8$)	0.6984	0.5094	5.07
Dodecane (C$_{12}$)	0.7450	1.3791	2.36
$x_{C_{12}}$	Heptane (C$_7$) + dodecane (C$_{12}$)		
0.2	0.6981	0.5157	4.89
0.4	0.7131	0.6772	4.07
0.6	0.7257	0.8662	3.37
0.8	0.7359	1.0858	2.80
$x_{C_{12}}$	Octane (C$_8$) + dodecane (C$_{12}$)		
0.2	0.7106	0.6353	4.31
0.4	0.7209	0.7703	3.66
0.6	0.7300	0.9477	3.12
0.8	0.7378	1.1383	2.69

Fig. 3. Influence of stirring power supplied upon the mass transfer coefficient in carbon dioxide/(octane + dodecane) system. (O) $x_{C_{12}} = 0$; (●) $x_{C_{12}} = 0.25$; (□) $x_{C_{12}} = 0.50$; (■) $x_{C_{12}} = 0.75$; (△) $x_{C_{12}} = 1$; $Q_g = 7.5$ L h^{-1}.

3.2.2. Alkanes mixtures

The effects produced by the operational variables previously analysed upon the absorption process employing binary mixtures were similar to the previous behaviours found in pure alkanes when they were employed as absorbent phases. Fig. 3 shows an example of the experimental results obtained, in relation to the effect produced by stirring power upon the gas/liquid mass transfer coefficient for different mixtures of octane and dodecane.

Since the influence of gas flow rate fed to the contactor upon the mass transfer velocity is quiet low in relation to the effect caused by the stirring rate (or stirring power) [18,22], this variable has not been studied in the present section.

Also, the effect caused by the binary mixture composition of liquid phases upon the mass transfer process has been analysed. The behaviour observed for the systems of carbon dioxide/alkanes mixtures has been summarized in Fig. 4. It is possible to observe in this figure that when the absorbent phase enriches the alkane with higher molecular weight, the gas mass transfer velocity to liquid phase decreases it value. The behaviours shown
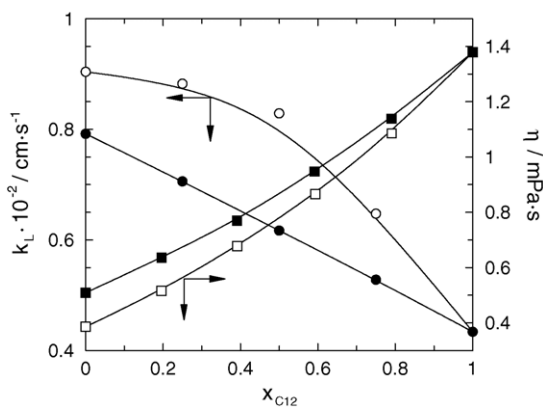

Fig. 4. Mass transfer coefficient for different compositions of alkanes mixtures. (O) and (□) *n*-heptane + *n*-dodecane system. (●) and (■) *n*-octane + *n*-dodecane system. $W = 250$ rpm; $Q_g = 7.5$ L h^{-1}.

in Fig. 4, in relation to the effect produced upon the physical properties of mixture composition, are similar for both systems. For the liquid phase formed for octane + dodecane, the values obtained for mass transfer coefficient were minor than from the other system. Using the experimental values shown in Fig. 4, it is possible to conclude that there is a certain deviation from linearity for both systems. Similar deviations were found when the effect of mixture composition upon the physical properties was analysed.

The obtained experimental results have been compared with the viscosity of alkanes mixtures. This physical property has been used to understand the absorption process due to the great importance of viscosity in mass transfer operations. Negative deviations were found for viscosity, whereas the opposite behaviour was found for mass transfer coefficient. An increase in the viscosity causes a decrease in mass transfer velocity of a gaseous component since the resistance to the transport is increased [22,23].

The obtained results for mass transfer coefficient correlate with the liquid phase viscosity variations. A similar behaviour was observed for the heptane + dodecane mixture analysed in the present study.

The effects caused by the operational variables and physical properties upon the mass transfer process have been analysed. The experimental values for absorption experiments have been employed to model the absorption process by means of calculating the liquid side mass transfer coefficient using fit equations for gas/liquid systems. These equations relate the transport coefficient in a direct or indirect way to the operational variables previously commented.

Useful expressions for concrete cases are equations based on the operational variables with an influence upon mass transfer processes. In the present paper, the stirring power supplied to the liquid phases and the absorbent nature, are variables with a high importance in modelling processes.

Eq. (7) shows the expression employed to fit the experimental data obtained for carbon dioxide absorption in linear alkanes mixtures.

$$k_L = 1.3 \cdot 10^{-8} \cdot P_{mo}^{0.30} \cdot \eta^{-0.81} \tag{7}$$

This equation has shown acceptable fits with low deviations when the two systems studied in the present paper are considered together (Fig. 5). The calculated values for fit parameters of Eq. (7) are similar to others obtained from researchers who have employed several systems [24].

On the other hand, numerous researchers have employed equations developed by means of dimensional analysis [25,26], based on dimensionless numbers where the variables previously studied are included. These equations allow the development of more general expressions applied to different gas/liquid systems and geometrical configurations. One of them has been employed in present paper to fit the experimental data is shown in Eq. (8).

$$Sh = C_1 \cdot Re^{0.8} \cdot Sc^{0.33} \tag{8}$$

where *Sh* is the modified Sherwood number, *Re* the Reynolds number and *Sc* is the Schmidt number. Employing this equation
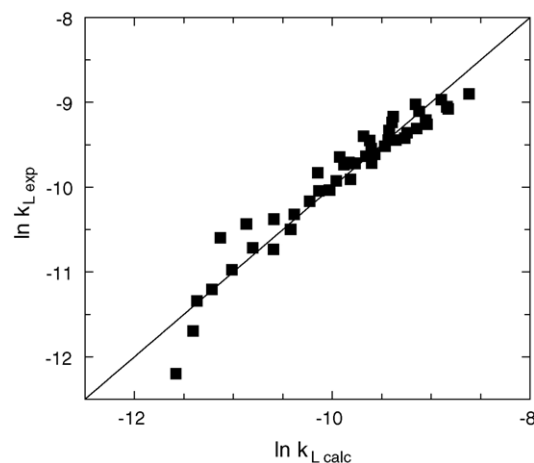

Fig. 5. Comparison between experimental and calculated values of mass transfer coefficient using Eq. (7).

to fit the data corresponding to mass transfer process using alkanes mixtures as liquid phases, the value for C_1 parameter was 0.0081.

To employ Eq. (8) it is necessary to know different properties of alkanes. Its mixtures, emphasized in density and viscosity, are listed in Table 1.

Also, in Sherwood and Schmidt numbers, one of the present variables is the gas diffusivity in the liquid phase, in this case, the diffusivity of carbon dioxide in alkanes and its mixtures. To determine this parameter we have chosen the bibliographic data supplied by Reid et al. [27]. These authors compiled values for carbon dioxide diffusivity in different organic liquids and developed a correlation between carbon dioxide diffusivity and liquid phase viscosity. The observed behaviour has a power trend and the correlation is shown in Eq. (9),

$$D = M \cdot \eta^m \tag{9}$$

where *D* is the gas diffusivity in the liquid phase, and η is the liquid phase viscosity. Using Eq. (9) and the *Q* and *q* parameters supplied by Reid et al., it is possible to estimate the carbon dioxide diffusivity in the linear alkanes and the mixtures employed in the present paper.

The comparison between the values of the mass transfer coefficient, which were determined experimentally, and those calculated employing Eq. (8) is shown in Fig. 6. The data plotted in this figure indicates that there is a high scattering in relation to the experimental and calculated data. When this equation was employed in a previous paper to fit the mass transfer coefficient using individual lineal alkanes as absorbent liquid phases, the results did not show this high scattering [18], and an acceptable fit was obtained. Our studies indicate that this equation contributes high deviations when it is used to calculate liquid side mass transfer coefficient.

A model developed by Lamont and Scott [28] was based on assumption that the mass transfer in the liquid is preponderantly affected by small scales of turbulent motions. The equation deduced by these authors has the expression shown in Eq. (10), and it has been employed in several cases for bubbling stirred
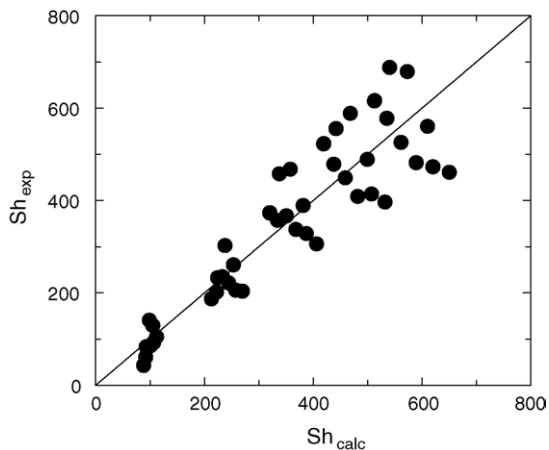
Fig. 6. Comparison between experimental and calculated values of Sherwood number using Eq. (8).

vessel,

$$k_{L} = C_{2} \cdot \left(\frac{P_{mo} \cdot \nu}{\rho} \right)^{1/4} \cdot \left(\frac{D}{\nu} \right)^{1/2} \tag{10}$$

where P_{mo} is the total specific power dissipated in the liquid phase, ν and ρ the kinematics viscosity and density of the liquid phase, D the diffusivity of gas phase in the liquid and c is an adjustable parameter. Fit parameter C_{2}, for the systems analysed in the present paper, takes a value of 0.018.

This equation (Eq. (10)) has been employed by several authors [29] to correlate experimental data of absorption corresponding to liquid side mass transfer coefficient, employing bubbling stirred vessels as contact devices. This equation takes into account important operation variables such as power supplied to the liquid, viscosity, density and gas diffusivity in the liquid. When the contactor is a bubbling reactor, the power must be calculated or determined as the sum of the contributions of the mechanical stirrer and the power supplied by the bubbling, due to the bubbles ascent along the liquid phase.

The comparison between experimental and calculated values of mass transfer coefficient using the model developed by
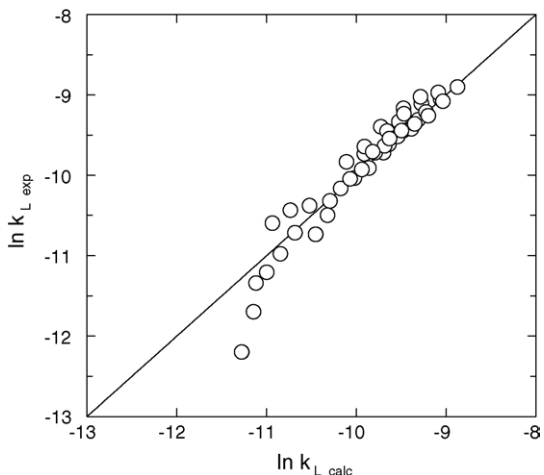

Fig. 7. Use of Lamont and Scott equation (Eq. (10)) to correlate mass transfer coefficients for the liquid phases employed in the present paper.

Lamont and Scott indicates that this equation allows the simulation of the absorption process for the systems employed in the present paper with good results (Fig. 7). This equation has been employed by several researchers [29] and our research group, obtaining good results.

4. Conclusions

Volumetric mass transfer coefficients corresponding to carbon dioxide absorption in binary mixtures of linear alkanes (heptane, octane and dodecane) have been determined using a flat surface gas/liquid contactor. The influence of the stirring rate applied and liquid phase nature upon absorption processes has been analysed in the present paper more specifically. The absorbent phases employed have different binary relations between the linear alkanes. The important effect of both variables upon the mass transfer velocity has been proved on the basis of obtained experimental data. The last point has been the use of three models to correlate the value of mass transfer coefficient at different values of operational variables. This study has shown that the correlation directly based on variables and the equation developed by Lamont and Scott satisfactorily fit the absorption experimental data for the studied systems.

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